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QUANTUM PISTON - QUANTUM PRESERVATION, SIMULATION AND TRANSFER IN OXIDE NANOSTRUCTURES

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QUANTUM PISTON - QUANTUM PRESERVATION, SIMULATION AND TRANSFER IN OXIDE NANOSTRUCTURES

Overview

This supersemi MURI project aimed to develop the unique properties of superconducting semiconductors to achieve Quantum Preservation, Simulation and Transfer in Oxide Nanostructures. We we have pursued three technical approaches:

1. Quantum Preservation: Using predicted topological properties of superconductors in two dimensions, we will develop ways for preserving quantum information in a way that is immune to a wide variety of decoherence mechanisms, thus paving the way for a scalable quantum information technology.
2. Quantum Simulation: The extreme nanoscale precision with which the $LaAlO_3/SrTiO_3$ interface can be gated will be used to program fundamental couplings at near-atomic scales and quantum simulation of "metasuperconductors".
3. Quantum Transfer: New mechanisms for the transfer of quantum information between long-lived localized states (nitrogen-vacancy centers) and delocalized states (superconducting resonators) will be developed.

Quantum Preservation

Both experimental and theoretical approaches were taken with regard to topological protection (quantum preservation) afforded by theoretically predicted Majorana fermion-based quantum bits. The theory of Majorana fermions has been extended by Nayak to allow them to exist in systems that lack long-range superconducting order. In addition, they have developed a model to accommodate both magnetism and superconductivity at the LAO/STO interface.

The main purposes of the theoretical activity supported by this MURI have been to (1) understand the causes and nature of the magnetic and superconducting ordering of the electrons at the LAO/STO interface and (2) determine whether Majorana zero modes might exist and be detected in nanowires 'drawn' at these interfaces by the University of Pittsburgh team.

In thinking about the possibility of Majorana zero modes in nanowires are LAO/STO interfaces, it became clear that one important feature of superconductivity in these wires is that it is quasi-one-dimensional (i.e. not induced by proximity to a 3D bulk superconductor, as in proposed semiconductor 'Majorana wires'). Therefore, we had to answer the question of whether Majorana zero modes could be supported by quasi-one-dimensional superconductors, which necessarily have superconducting order with power-law decay, not true long-ranged order. In the published paper listed above, we showed that such a superconductor could (under the right circumstances) support Majorana zero modes.

Quantum Simulation

Important experimental advances have brought us close to a regime in which quantum simulation will be possible. True quantum simulation requires a deep (first-principles-level) understanding of the host material that is performing the simulation. A lot of the research that was conducted involves basic characterization (quantum transport) in LAO/STO nanostructures that have not been patterned in to

artificial lattices. We have however performed preliminary results (presented at conferences) that show that this approach is both feasible and highly promising.

Quantum Transfer

The ability to transfer quantum information from one physical embodiment to another represents an essential building block in any quantum architecture. Work in this area has mainly involved diamond, a superconducting semiconductor with long-lived defect complexes (nitrogen-vacancy or NV centers). A central goal of the project was to integrate diamond with LAO/STO heterostructures. This approach proved to be highly challenging, if technically feasible. Structures have been shared with Eom, Levy, Schlom and Pan. Eom and Schlom have also grown epitaxial LAO/STO heterostructures on cubic SiC/Si substrates by PLD with high pressure RHEED.

Cross-Cutting Activities

A number of cross-cutting activities (growth of superconducting-semiconductor structures, atomic-scale characterization, electrical and optical experiments) will take place, serving multiple tasks. The successful development of new quantum technologies from this research will enable new and powerful types of quantum speedups of computational tasks, including solutions to large systems of linear equations, fully quantum-mechanical simulation of materials, and interoperability among quantum platforms.

Quantum Transport in LAO/STO

A major focus in Quantum Preservation has been the development of superconducting nanowires and possible topological phases that could be used to support Majorana fermions. The first fully superconducting nanowires were demonstrated. The normal state of these structures is unusual in that the four-terminal resistance is independent of the length of the channel and always of order h/e^2. In addition, some unusual nonlocal Hall effects have been observed which imply an unusually long coherence length in the system. We also observe greatly enhanced mobility that extends up to room temperature.

Rashba Spin-Orbit Coupling

We study origin of Rashba spin-orbit interaction at SrTiO3 surfaces and LaAlO3/SrTiO3 interfaces by considering the interplay between atomic spin-orbit coupling and inversion asymmetry at the surface or interface. We show that in a simple tight-binding model involving 3d t2g bands of Ti ions, the induced spin-orbit coupling in the dxz and dyz bands is cubic in momentum whereas the spin-orbit interaction in the dxy band has linear momentum dependence. We also find that the **spin-orbit interaction in one-dimensional channels at LaAlO3/SrTiO3 interfaces is linear in momentum for all bands.** We discuss implications of our results for transport experiments on SrTiO3 surfaces and LaAlO3/SrTiO3 interfaces. In particular, we analyze the effect of a given spin-orbit interaction term on magnetotransport of LaAlO3/SrTiO3 by calculating weak antilocalization corrections to the conductance and to universal conductance fluctuations.

1D Superconductivity in LAO/STO

We report quasi-1D superconductivity at the interface of LaAlO3 and SrTiO3. The material system and nanostructure fabrication method supply a **new platform for superconducting nanoelectronics**. Nanostructures having line widths w~10 nm are formed from the parent two-dimensional electron liquid using conductive atomic force microscope lithography. Nanowire cross-sections are small

compared to the superconducting coherence length in LaAlO3/SrTiO3, placing them in the quasi-1D regime. Broad superconducting transitions versus temperature and finite resistances in the superconducting state well below Tc ≈200 mK are observed, suggesting the presence of fluctuation- and heating-induced resistance. The superconducting resistances and V–I characteristics are tunable through the use of a back gate. Four-terminal resistances in the superconducting state show an unusual dependence on the current path, varying by as much as an order of magnitude. This new technology, i.e., the ability to 'write' gate-tunable superconducting nanostructures on an insulating LaAlO3/SrTiO3 'canvas', opens possibilities for the development of new families of reconfigurable superconducting nanoelectronics.

Nonlocal transport in LAO/STO

The oxide heterostructure LaAlO3/SrTiO3 supports a two-dimensional electron liquid with a variety of competing phases, including magnetism, superconductivity, and weak antilocalization because of Rashba spin-orbit coupling. Further confinement of this two-dimensional electron liquid to the quasi-one-dimensional regime can provide insight into the underlying physics of this system and reveal new behavior. Here, we describe magnetotransport experiments on narrow LaAlO3/SrTiO3 structures created by a conductive atomic force microscope lithography technique. Four-terminal local-transport **measurements on Hall bar structures about 10 nm wide yield longitudinal resistances that are comparable to the resistance quantum h/e2 and independent of the channel length.** Large nonlocal resistances (as large as 10^4 Ω) are observed in some but not all structures with separations between current and voltage that are large compared to the two-dimensional mean-free path. The nonlocal transport is strongly suppressed by the onset of superconductivity below about 200 mK. The origin of these anomalous transport signatures is not understood, but may arise from coherent transport defined by strong spin-orbit coupling and/or magnetic interactions.

Theory of magnetism and superconductivity at LAO/STO

We formulated a model for magnetic and superconducting ordering in one-dimensional nanostructures at LaAlO3/SrTiO3 interfaces containing both localized magnetic moments and itinerant electrons. Although these both originate in Ti 3d orbitals, the former may be due to electrons more tightly bound to the interface, while the latter are extended over several layers. Only the latter contribute significantly to metallic conduction and superconductivity. In our model, the interplay between the two types of electrons, which is argued to be ferromagnetic, combined with strong spin-orbit coupling of the itinerant electrons, leads to magnetic ordering. Furthermore, we propose a model for interfacial superconductivity, consisting of random superconducting grains in the bulk SrTiO3 driven, via coupling to the interface conduction band, towards long-ranged or quasi-long-ranged order. Most interestingly, the **magnetic order and strong spin-orbit coupling can lead in this manner to unconventional interfacial superconductivity, yielding a possible realization of Majorana physics**.

Effect of ferroelastic domain walls

The ability to control materials properties through interface engineering is demonstrated by the appearance of conductivity at the interface of certain insulators, most famously the {001} interface of the band insulators LaAlO3 and TiO2-terminated SrTiO3 (STO). Transport and other measurements in this system show a plethora of diverse physical phenomena. To better understand the interface conductivity, we used scanning superconducting quantum interference device microscopy to image the magnetic field locally generated by current in an interface. At low temperature, **we found that the current flowed in conductive narrow paths oriented along the crystallographic axes**, embedded in a

less conductive background. The configuration of these paths changed on thermal cycling above the STO cubic-to-tetragonal structural transition temperature, implying that the local conductivity is strongly modified by the STO tetragonal domain structure. The interplay between substrate domains and the interface provides an additional mechanism for understanding and controlling the behaviour of heterostructures.

Nonlocal transport

Effects from **nonequilibrium superconductivity** play a major role in the physics of superconducting nanoelectronics. Notably, charge imbalance arising from the point at which the superconducting device contacts normal-metal leads is prevalent, particularly in reduced dimensions. We investigate nonlocal transport signatures in quasi-1D nanostructures formed at the LaAlO3/SrTiO3 interface. The nonlocal resistances correlate with the bias, magnetic field, and back gate dependence of the superconducting state. We attribute these signatures to charge imbalance or spin-dependent excitations. Understanding and control over these effects are important for further development of superconducting nanoelectronics in this material system, including the ability to probe the interaction of superconductivity and other rich physics in LaAlO3/SrTiO3 on the nanoscale.

Electron Pairing Without Superconductivity

Strontium titanate (SrTiO3) is the first and best known superconducting semiconductor. It exhibits an extremely low carrier density threshold for superconductivity, and possesses a phase diagram similar to that of high-temperature superconductors—two factors that suggest an unconventional pairing mechanism. Despite sustained interest for 50 years, direct experimental insight into the nature of electron pairing in SrTiO3 has remained elusive. Here we perform transport experiments with nanowire-based single-electron transistors at the interface between SrTiO3 and a thin layer of lanthanum aluminate, LaAlO3. Electrostatic gating reveals a series of two-electron conductance resonances—paired electron states—that bifurcate above a critical pairing field Bp of about 1–4 tesla, an order of magnitude larger than the superconducting critical magnetic field. For magnetic fields below Bp, these resonances are insensitive to the applied magnetic field; for fields in excess of Bp, the resonances exhibit a linear Zeeman-like energy splitting. Electron pairing is stable at temperatures as high as 900 millikelvin, well above the superconducting transition temperature (about 300 millikelvin). **These experiments demonstrate the existence of a robust electronic phase in which electrons pair without forming a superconducting state.** Key experimental signatures are captured by a model involving an attractive Hubbard interaction that describes real-space electron pairing as a precursor to superconductivity.

Magnetism in LAO/STO

Room temperature electronically controlled ferromagnetism

Reports of emergent conductivity, superconductivity and magnetism have helped to fuel intense interest in the rich physics and technological potential of complex-oxide interfaces. Here we employ magnetic force microscopy to search for room-temperature magnetism in the well-studied LaAlO3/SrTiO3 system. Using electrical top gating to control the electron density at the oxide interface, **we directly observe the emergence of an in-plane ferromagnetic phase as electrons are depleted from the interface.** Itinerant electrons that are reintroduced into the interface align antiferromagnetically with the magnetization at first screening and then destabilizing it as the conductive regime is approached. Repeated cycling of the gate voltage results in new, uncorrelated magnetic patterns. This newfound

control over emergent magnetism at the interface between two non-magnetic oxides portends a number of important technological applications.

Growth of LAO/STO Heterostructures

Much of what was proposed for this project relates to $LaAlO_3/SrTiO_3$ (LAO/STO) heterostructures, which are grown primarily in the Eom group. They have provided structures that have been shared throughout the team, including Levy, Awschalom, Schlom and Pan. One key scientific finding of Eom relates to the effect of **different lattice constant single crystal substrates to produce LAO/STO interfaces with controlled levels of biaxial epitaxial strain**. Tensile strained $SrTiO_3$ destroys the conducting 2DEG, while compressively strained $SrTiO_3$ retains the 2DEG, but with a carrier concentration reduced in comparison to the unstrained LAO/STO interface. The critical $LaAlO_3$ overlayer thickness for 2DEG formation increases with $SrTiO_3$ compressive strain. First-principles calculations suggest that a strain-induced electric polarization in the $SrTiO_3$ layer is responsible for this behavior. It is directed away from the interface and hence creates a negative polarization charge opposing that of the polar $LaAlO_3$ layer. This both increases the critical thickness of the $LaAlO_3$ layer, and reduces carrier concentration above the critical thickness, in agreement with our experimental results. Our findings suggest that epitaxial strain and induced polarization can be used to tailor 2DEGs properties of the LAO/STO heterointerface.

Eom's group **demonstrated $LaAlO_3/SrTiO_3$ heterointerfaces grown by 90° off-axis sputtering** which allows uniform films over a large area. The electrical transport properties of the $LaAlO_3/SrTiO_3$ heterointerface are similar to those grown by pulsed laser deposition. We also demonstrated room-temperature conductive probe-based switching of quasi-one-dimensional structures. This work demonstrates that a scalable growth process can be used to create the two-dimensional electron gas system at oxide heterointerfaces.

Diamond-based Heterostructures and N-V Centers

Our MURI team was able to integrate **phase-pure EuO on both single-crystal diamond** and on epitaxial diamond films grown on silicon utilizing reactive molecular-beam epitaxy. The epitaxial orientation relationship is (001) EuO ‖ (001) diamond and [110] EuO ‖ [100] diamond. The EuO layer is nominally unstrained and ferromagnetic with a transition temperature of 68 ± 2 K and a saturation magnetization of 5.5 ± 0.1 Bohr magnetons per europium ion on the single-crystal diamond, and a transition temperature of 67 ± 2 K and a saturation magnetization of 2.1 ± 0.1 Bohr magnetons per europium ion on the epitaxial diamond film.

The properties of **NV centers** were developed for a number of **quantum measurement** challenges. Extension of nuclear magnetic resonance (NMR) to nanoscale samples has been a longstanding challenge because of the insensitivity of conventional detection methods. We demonstrated the use of an individual, near-surface nitrogen-vacancy (NV) center in diamond as a sensor to detect proton NMR in an organic sample located external to the diamond. Using a combination of electron spin echoes and proton spin manipulation, we showed that the NV center senses the nanotesla field fluctuations from the protons, enabling both time-domain and spectroscopic NMR measurements on the nanometer scale.

Theoretical calculations and analysis of the vibronic structure of the spin-triplet optical transition in diamond nitrogen-vacancy (NV) centres were carried out. The electronic structure of the defect is

described using accurate first-principles methods based on hybrid functionals. We devise a computational methodology to determine the coupling between electrons and phonons during an optical transition in the dilute limit. As a result, our approach yields a smooth spectral function of electron–phonon coupling and includes both quasi-localized and bulk phonons on equal footings. The luminescence lineshape is determined via the generating function approach. We obtain a highly accurate description of the luminescence band, including all key parameters such as the Huang–Rhys factor, the Debye–Waller factor, and the frequency of the dominant phonon mode. More importantly, our work provides insight into the vibrational structure of NV centres, in particular the role of local modes and vibrational resonances. In particular, we find that the pronounced mode at 65 meV is a vibrational resonance, and we quantify localization properties of this mode. These excellent results for the benchmark diamond (NV) centre provide confidence that the procedure can be applied to other defects, including alternative systems that are being considered for applications in quantum information processing.

Electric field noise from **surface charge fluctuations can be a significant source of spin decoherence** for near-surface nitrogen-vacancy (NV) centers in diamond. This conclusion is based on the increase in spin coherence observed when the diamond surface is covered with high-dielectric-constant liquids, such as glycerol. Double-resonance experiments show that improved coherence occurs even though the coupling to nearby electron spins is unchanged when the liquid is applied. Multipulse spin-echo experiments reveal the effect of glycerol on the spectrum of NV frequency noise.

Silicon Carbide Divacancy Centers

A new discovery made by Awschalom during the MURI relates to the development of long-lived spin states in SiC. Schlom has demonstrated epitaxial growth of $SrTiO_3$ (111) on SiC, and hence this new hybrid system constitutes a new focus for our MURI team, but falling within the original goals of the project. Alternative solid state materials for hosting quantum information processing are explored. Multiple polytypes of SiC have been identified as viable candidates.

Crystal defects can confine isolated electronic spins and are promising **candidates for solid-state quantum information**. Alongside research focusing on nitrogen-vacancy centres in diamond, an alternative strategy seeks to identify new spin systems with an expanded set of technological capabilities, a materials-driven approach that could ultimately lead to 'designer' spins with tailored properties. Here we show that the 4H, 6H and 3C polytypes of SiC all host coherent and optically addressable defect spin states, including states in all three with room-temperature quantum coherence. The prevalence of this spin coherence shows that crystal polymorphism can be a degree of freedom for engineering spin qubits. Long spin coherence times allow us to use double electron–electron resonance to measure magnetic dipole interactions between spin ensembles in inequivalent lattice sites of the same crystal. Together with the distinct optical and spin transition energies of such inequivalent states, these interactions provide a route to dipole-coupled networks of separately addressable spins.

It was demonstrated that the spin of **optically addressable point defects can be coherently driven with ac electric fields**. Based on magnetic-dipole forbidden spin transitions, this scheme enables spatially confined spin control, the imaging of GHz-frequency electric fields, and the characterization of defect spin multiplicity. We control defect ensembles in SiC, but our methods apply to spin systems in many semiconductors, including the diamond nitrogen-vacancy center. Electrically driven spin resonance offers a viable route towards scalable quantum control of electron spins in a dense array.

Multipulse magnetometry exploits all three magnetic sublevels of the S=1 nitrogen-vacancy center in diamond to achieve enhanced magnetic field sensitivity. Based on dual frequency microwave pulsing, the scheme is twice as sensitive to ac magnetic fields as conventional two-level magnetometry. We derive the spin evolution operator for dual frequency microwave excitation and show its effectiveness for double-quantum state swaps. Using multipulse sequences of up to 128 pulses under optimized conditions, we show enhancement of the SNR by up to a factor of 2 in detecting NMR statistical signals, with a 4× enhancement theoretically possible.

We demonstrate optically pumped dynamic nuclear polarization of Si29 nuclear spins that are strongly coupled to paramagnetic color centers in 4H- and 6H-SiC. The 99%±1% degree of polarization that we observe at room temperature corresponds to an effective nuclear temperature of 5 µK. By combining ab initio theory with the experimental identification of the color centers' optically excited states, we quantitatively model how the polarization derives from hyperfine-mediated level anticrossings. These results lay a foundation for SiC-based quantum memories, nuclear gyroscopes, and hyperpolarized probes for magnetic resonance imaging.

Characterization

High-resolution TEM measurements of LAO/STO heterostuctures have been performed by Pan, and they have helped to understand the role of interdiffusion across the LAO/STO interface as well as the electronic polarity of this interface. Determination of the microstructures and interfacial properties of LaAlO3 / SrTiO3 device structures by transmission electron microscopy (TEM) in combination with focused ion beam TEM specimen preparation. We found that Nb in prelayer diffused into Au electrode and that the LAO/STO interface, where the 2DEG forms, is directly contacting to the prelayer but not the Au electrode. These studies provide important feedback to the nanofabrication of the device structures. Determination of microstructures and interfacial atomic structure of LaAlO3 / SrTiO3 / SiC heterostructures grown by molecular beam epitaxy. It was found that the SrTiO3 grows epitaxially on the (001) SiC substrate, showing a rough surface and a high density of vertical twin boundaries due to the existence of two different twinning variants. As a result, the LaAlO3 over-layer has a poor microstructure and a rough interface to the SrTiO3 layer.

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Archival Publications (published) during reporting period:

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2. New discoveries, inventions, or patent disclosures:

Do you have any discoveries, inventions, or patent disclosures to report for this period?

Yes

Please describe and include any notable dates

UPITT Ref. No.: 03092
Foley Ref. No.: 076333-0683
Title: Oxide Interface Displaying Electronically Controllable Ferromagnetism
Assigned Application No.: 14/801,410
Filed: July 16, 2015

Do you plan to pursue a claim for personal or organizational intellectual property?

No

Changes in research objectives (if any):

Change in AFOSR Program Manager, if any:

Extensions granted or milestones slipped, if any:

One NCE after Period 5 was requested and granted.

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, $K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
Total			

Report Document

Report Document - Text Analysis

Report Document - Text Analysis

Appendix Documents

2. Thank You

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